

IN THE U.S. PATENT AND TRADEMARK OFFICE

Applicants: Akira NAKABAYASHI et al.

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For: PHOTOCATALYST COMPOSITION

Art Unit: 1755

Examiner: JENNINE M. BROWN

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DECLARATION UNDER 37 C.F.R. 1.132

I, the undersigned, Akira NAKABAYASHI, a Japanese citizen, residing at 3-36-3-323, Ookubo, Konan-ku, Yokohama-shi Kanagawa 233-0007 Japan, hereby declare and state that:

I took Doctor Course at the Department of Electronic Chemistry, the Interdisciplinary Graduate School of Science and Engineering, Tokyo Institute of Technology, and I was graduated therefrom in March 2005.

I entered Asahi Kasei Kogyo Kabushiki Kaisha in April 1987.

I was engaged in research on polymer chemistry in connection with the research and development of coating materi-

als from April 1987 to March 1998.

I have been engaged in the research and development of photocatalysts and photocatalyst-containing materials from April 1998 to date.

I am one of the applicants of the above-identified application and I am well familiar with the present case.

I have read and understood the Office Action dated January 24, 2006 and the references cited therein.

I carried out Examples 1 to 7 and Comparative Examples 1 to 5 of the present application, and the results are as described on pages 117 to 143 of the specification of the present application.

I have made observations, with reference to Example 1 and Comparative Examples 1 and 3 of the present specification, to show that the binder component (B) (comprising the phenyl group-containing silicone) (used in combination with the modified photocatalyst particles (A)) as recited in claim 1 of the present application is critical for exhibiting the self-stratifying or self-gradating property, which leads to the excellent effects of the photocatalyst composition of the present invention, i.e., the effects that it can be used to

produce a functional composite which is advantageous in that it is free from deterioration occurring at the interface between the substrate and the photocatalyst-containing film and free from deterioration of the binder component in the photocatalyst-containing film, and in that it exhibits an excellent balance of hardness and flexibility (impact resistance), and it has excellent durability such that the surface of the functional composite exhibits, by light irradiation, a water contact characteristic (i.e., hydrophilicity or hydrophobicity) and/or maintains such photocatalyst activity for a long time. The method and results are as described in a paper attached hereto and marked "Exhibit".

From the results of the Exhibit, it can be fairly concluded:

that the data of Example 1 and Comparative Examples 1 and 3 of the present specification are as summarized in Table A below:

Table A

		<b>Example 1</b> (present invention)	<b>Comparative Example 1</b>	<b>Comparative Example 3</b>
photocatalyst composition	Type of photocatalyst	<b>modified</b> photocatalyst	<u>unmodified</u> photocatalyst	<b>modified</b> photocatalyst
	Type of binder component	phenyl group-containing binder component (B-1)	phenyl group-containing binder component (B-1)	binder component (B-3) containing <u>no</u> phenyl group
self-stratifying or self-gradating property		<b>exhibited</b>	<b>not exhibited</b>	<b>not exhibited</b>
photocatalyst activity *)		<b>excellent</b> (symbol: ⊙)	<b>poor</b> (symbol: ×)	<b>poor</b> (symbol: ×)
weathering resistance		<b>excellent</b>  (gloss retention value was as high as 98 %)	<b>very poor</b>  (gloss retention value was as low as 10 % or less, and a chalking was observed)	<b>extremely poor</b>  (the film <u>peeled off</u> from the substrate)

that, as seen from Table A above, in **Example 1** of the present application (using the **binder component (B)** (comprising the **phenyl group-containing silicone**) in combination with the **modified photocatalyst particles (A)**, as required by **claim 1** of the present application), it is found that the

self-stratifying or self-gradating property is exhibited; the photocatalyst activity is excellent (evaluation symbol: ⊙); and the gloss retention value is as high as 98 %, indicating excellent weathering resistance;

that, by contrast, in Comparative Example 1 of the present application (which does not satisfy the component (A) requirement of claim 1 of the present application), it is found that the self-stratifying or self-gradating property is not exhibited; the photocatalyst activity is poor (evaluation symbol: ×); and the gloss retention value was as low as 10 % or less, and a chalking was observed, indicating very poor weathering resistance; and

that likewise, in Comparative Example 3 of the present application (which does not satisfy the component (B) requirement of claim 1 of the present application), it is found that the self-stratifying or self-gradating property is not exhibited; the photocatalyst activity is poor (evaluation symbol: ×); and when the weathering resistance test is performed using a Dewpanel light control weather meter, the film peeled off from the substrate, indicating extremely poor weathering resistance; and

that, from the above, it is quite apparent that the binder component (B) (comprising the phenyl group-containing silicone) (used in combination with the modified photocatalyst particles (A)) as recited in claim 1 of the present ap-

plication is critical for exhibiting the self-stratifying or self-gradating property, which leads to the excellent effects of the photocatalyst composition of the present invention.

The undersigned petitioner declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: July 18, 2006

Akira Nakabayashi  
Akira NAKABAYASHI

Observations, with reference to **Example 1** and **Comparative Examples 1 and 3** of the present specification, to show that the **binder component (B)** (comprising the **phenyl group-containing silicone**) (used in combination with the **modified photocatalyst particles (A)**) as recited in claim 1 of the present application is **critical** for exhibiting the **self-stratifying or self-gradating property**, which leads to the excellent effects of the **photocatalyst composition of the present invention**

1. Object:

The object of the observations is to show that the **binder component (B)** (comprising the **phenyl group-containing silicone**) (used in combination with the **modified photocatalyst particles (A)**) as recited in claim 1 of the present application is **critical** for exhibiting the **self-stratifying or self-gradating property**, which leads to the excellent effects of the photocatalyst composition of the present invention, i.e., the effects that it can be used to produce a functional composite which is advantageous in that it is **free from deterioration** occurring at the interface between the substrate and the photocatalyst-containing film and free from deterioration of the binder component in the photocatalyst-

containing film, and in that it exhibits an excellent balance of hardness and flexibility (impact resistance), and it has **excellent durability** such that the surface of the functional composite exhibits, by light irradiation, a water contact characteristic (i.e., hydrophilicity or hydrophobicity) and/or maintains such photocatalyst activity for a long time.

## 2. Observations:

### (2-1)

In **Example 1** of the present specification (which is described at page 117, line 22 to page 121, line 21 of the present specification), simply stated, the following experiments were performed.

20 g of the **modified** photocatalyst organosol (A-1) and 68 g of the solution of **phenyl group**-containing binder component (B-1) (which was produced in Reference Example 5) were used to obtain photocatalyst composition (C-1), which **satisfies** the requirements of **claim 1** of the present application.

An aluminum plate having a size of 50 mm × 60 mm and a thickness of 1 mm (JIS, H, 4000 (Al050P)) was spray-coated with nax mightylac GII (color: white) (trade name of an acrylic urethane resin coating composition (two-pack coating composition)), followed by drying at room temperature for 3 days. Then, photocatalyst composition (C-1) obtained above was spray-coated onto the thus obtained acrylic urethane



coating formed on the aluminum plate so that a coating of photocatalyst composition (C-1) having a thickness of 2  $\mu\text{m}$  was formed and, then, the spray-coated photocatalyst composition (C-1) was dried at room temperature for 1 hour, followed by heating at 150 °C for 30 minutes, to thereby obtain test plate (D-1) having a photocatalyst-containing film.

A cross-section of the obtained test plate (D-1) having a photocatalyst-containing film was analyzed by TEM (transmission electron microscope) observation, and the results of the TEM analysis show that the modified photocatalyst particles were not present at the interface between the photocatalyst-containing film and the substrate which is the acrylic urethane coating (see Figs. 3(a) and 3(b) of the present application); that is, Figs. 3(a) and 3(b) of the present application clearly show that the **self-stratifying or self-gradating property** is exhibited by the photocatalyst composition used in Example 1.

In addition, the photocatalyst activity of the photocatalyst-containing film was evaluated and it was found that the **photocatalyst activity** was excellent (evaluation symbol: ©). The method for evaluating the photocatalyst activity is described at page 105, lines 6 to 23 of the present specification. Simply stated, the evaluation method is a method in which the photocatalyst activity to decompose methylene blue is tested and evaluated using the following 3 criteria:

©: methylene blue is completely decomposed;  
Δ: blue color of methylene blue slightly remains; and  
x: almost no decomposition of methylene blue is observed.

Further, an atmospheric exposure test was performed (for 1,000 hours) by a Dewpanel light control weather meter to determine the gloss retention value of test plate (D-1). The gloss retention value was as high as 98 %, indicating that test plate (D-1) had excellent weathering resistance.

(2-2)

In **Comparative Example 1** of the present specification (which is described at page 124, line 2 to page 125, line 5 of the present specification), a photocatalyst composition was produced in substantially the same manner as in Example 1 except that 10 g of unmodified photocatalyst organosol (TKS-251) was used instead of 20 g of modified photocatalyst organosol (A-1), thereby obtaining photocatalyst composition (C-2), which does not satisfy the requirements of **claim 1** of the present application.

Using the produced photocatalyst composition (C-2), a test plate was produced in the same manner as in Example 1, thereby obtaining test plate (D-3) having a photocatalyst-containing film (having a titanium oxide content which is the same as in Example 1).

The photocatalyst activity of the photocatalyst-containing film was evaluated and it was found that the photocatalyst activity was poor (evaluation symbol: ×).

Further, an atmospheric exposure test was performed (for 200 hours) by a Dewpanel light control weather meter to determine the gloss retention value of test plate (D-3). The gloss retention value was as low as 10 % or less, and a chalking was observed, indicating that test plate (D-3) had very poor weathering resistance.

Such poor results of the evaluation clearly show that the photocatalyst composition used in Comparative Example 1 (using an unmodified photocatalyst) cannot exhibit the self-stratifying or self-gradating property.

(2-3)

In Comparative Example 3 of the present specification (which is described at page 126, line 20 to page 127, line 24 of the present specification), a photocatalyst composition was produced in substantially the same manner as in Example 1 except that 68 g of the solution of binder component (B-3) containing no phenyl group and containing an alkyl group (which was produced in Reference Example 7) was used instead of binder component (B-1) (produced in Reference Example 5), thereby obtaining photocatalyst composition (C-3), which does not satisfy the requirements of claim 1 of the present appli-

cation.

Using the produced photocatalyst composition (C-3), a test plate was produced in the same manner as in Example 1, thereby obtaining test plate (D-5) having a photocatalyst-containing film (having a titanium oxide content which is the same as in Example 1).

The photocatalyst activity of the photocatalyst-containing film was evaluated and it was found that the photocatalyst activity was poor (evaluation symbol: ×).

Further, an atmospheric exposure test was performed (for 200 hours) by a Dewpanel light control weather meter. As a result, the film peeled off from the substrate, indicating that test plate (D-5) had extremely poor weathering resistance.

Such poor results of the evaluation clearly show that the photocatalyst composition used in Comparative Example 3 (using a binder component (B-3) containing no phenyl group and containing an alkyl group) cannot exhibit the self-stratifying or self-gradating property.

(2-4)

For easier understanding, the above-described data of Example 1 and Comparative Examples 1 and 3 of the present specification are summarized in Table A below.

Table A

		<b>Example 1</b> (present invention)	<b>Comparative Example 1</b>	<b>Comparative Example 3</b>
photocatalyst composition	Type of photocatalyst	<b>modified</b> photocatalyst	<b><u>unmodified</u></b> photocatalyst	<b>modified</b> photocatalyst
	Type of binder component	<b>phenyl group-containing</b> binder component (B-1)	<b>phenyl group-containing</b> binder component (B-1)	binder component (B-3) containing <b><u>no</u></b> <b><u>phenyl group</u></b>
self-stratifying or self-gradating property		<b>exhibited</b>	<b>not exhibited</b>	<b>not exhibited</b>
photocatalyst activity *)		<b>excellent</b> (symbol: ⊙)	<b>poor</b> (symbol: ×)	<b>poor</b> (symbol: ×)
weathering resistance		<b>excellent</b>  (gloss retention value was as high as 98 %)	<b>very poor</b>  (gloss retention value was as low as 10 % or less, and a <b>chalking</b> was observed)	<b>extremely poor</b>  (the film <b><u>peeled off</u></b> from the substrate)

Note \*): As mentioned above, simply stated, the method for evaluating the photocatalyst activity (described at page 105, lines 6 to 23 of the present specification) is a method in which the photocatalyst activity to decompose methylene blue is tested and evaluated using the following 3 criteria:

⊙: methylene blue is completely decomposed;

Δ: blue color of methylene blue slightly remains; and

×: almost no decomposition of methylene blue is observed.

### 3. Conclusion:

Thus, in **Example 1** of the present application (using the **binder component (B)** (comprising the **phenyl group-containing silicone**) in combination with the **modified photocatalyst particles (A)**, as required by **claim 1** of the present application), it is found that the **self-stratifying or self-gradating property** is exhibited; the **photocatalyst activity** is **excellent** (evaluation symbol: ©); and the gloss retention value is as high as **98 %**, indicating **excellent** weathering resistance.

By contrast, In **Comparative Example 1** of the present application (which does not satisfy the component (A) requirement of **claim 1** of the present application), it is found that the self-stratifying or self-gradating property is not exhibited; the **photocatalyst activity** is poor (evaluation symbol: ×); and the gloss retention value was as low as 10 % or less, and a chalking was observed, indicating very poor weathering resistance.

Likewise, in **Comparative Example 3** of the present application (which does not satisfy the component (B) requirement of **claim 1** of the present application), it is found that the self-stratifying or self-gradating property is not exhibited;

the photocatalyst activity is poor (evaluation symbol: ×); and when the weathering resistance test is performed using a Dewpanel light control weather meter, the film peeled off from the substrate, indicating extremely poor weathering resistance.

From the above, it is quite apparent that the binder component (B) (comprising the phenyl group-containing silicone) (used in combination with the modified photocatalyst particles (A)) as recited in claim 1 of the present application is critical for exhibiting the self-stratifying or self-gradating property, which leads to the excellent effects of the photocatalyst composition of the present invention.